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ABSTRACT - *Contrary to the behavior of swelling of f.c.c. Fe-15Cr-16Ni and Fe-15Cr-16Ni-0.25Ti alloys irradiated together in the same FFTF-MOTA experiment, Fe-15Cr-16Ti-0.25Ti-0.05C does not exhibit a dependence of swelling on dpa rate at ~400ºC. The transient regime of swelling is prolonged by carbon addition, however. Addition of boron to the carbon-doped alloy decreases the swelling somewhat but does not restore the sensitivity to dpa rate. It appears that the primary influence of boron is chemical in nature, probably associated with boron's impact on the behavior of carbon. Boron's role as a source of helium is thought to be secondary.*

INTRODUCTION

It is now recognized that the austenitic internal components of PWRs will experience some degree of void swelling and that the effect of the lower atomic displacement rates in PWRs compared to those in fast reactors causes some uncertainty in the application of fast reactor data to PWRs. Thus, it is important to study the effects of dpa rate on swelling and its synergisms with important material and environmental variables.

In an earlier report it was shown that two simple, annealed austenitic alloys, ternary Fe-15Cr-16Ni and quaternary Fe-15Cr-16Ni-0.25Ti, when irradiated in FFTF-MOTA at ~400ºC over a wide range of dpa rates, exhibited a very strong influence of dpa rate on void swelling (1). While the steady state swelling rate of ~1%/dpa was unaffected by dpa rate, the transient regime was strongly affected, with a progressive shortening of the transient duration as the dpa rate decreased, as shown in Figure 1. This counterintuitive dependence was shown to be mirrored in other previously published experiments conducted on more complex commercial alloys. (2-4). It was also shown that the addition of 500 appm boron to the ternary and quaternary alloys had very little effect on swelling.

Also contained in the FFTF-MOTA experiment was Fe-15Cr-16Ti-0.25Ti-0.05C, also in the annealed condition. Boron-doped variants of both Fe-15Cr-16Ni and Fe-15Cr-16Ni-0.25Ti-0.05C were also included. Boron

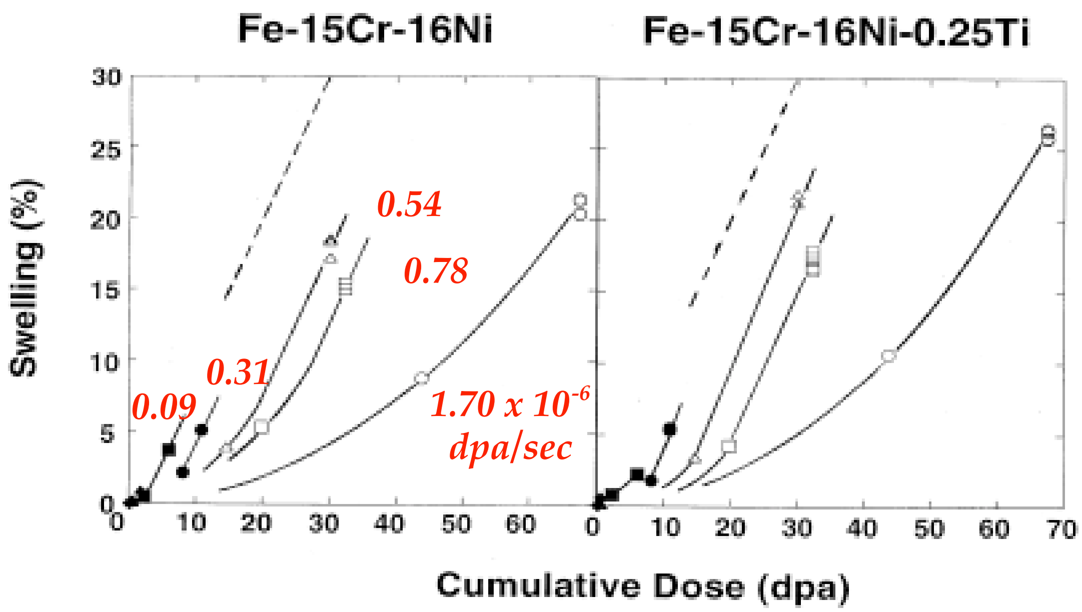

Figure 1 Swelling of simple model f.c.c. Fe-Cr-Ni alloys in FFTF-MOTA at ~400ºC, as observed by Okita and coworkers (1), showing that the transient regime of swelling increases progressively as the dpa rate increases.

additions not only change the matrix composition but also the helium generation rate. In Phase 2 of this experiment the possibly synergistic effects are studied of several important variables on void swelling, namely dpa rate, minor element composition and helium.

EXPERIMENTAL DETAILS

Relatively pure Fe-15Cr-16Ni, Fe-15Cr-16Ni-0.25Ti and Fe-15Cr-16Ni-0.25Ti-0.05C (at %) with no added solute were prepared by arc melting from high purity Fe, Ni, Cr and Ti. The alloys was rolled to sheets of 0.25 mm thickness, cut into 3 mm disks and annealed for 30 minutes at 1050°C in high vacuum. Also included in some sets of specimens were Fe-15Cr-16Ni-500 appm natural boron, and/or Fe-15Cr-16Ni-0.25Ti-0.05C with boron levels of 100, 500 and 2500 appm boron, all prepared using the same procedures.

Two sets of identical specimens are placed in sealed, helium-filled packets at each of seven different capsule positions of the Materials Open Test Assembly (MOTA), ranging from below the core to above the core of the Fast Flux Test Facility (FFTF). The packets in general contained four identical specimens of each of the two alloys. The various alloys were located side-by-side in the same packet. Two or more identically-loaded packets were placed in each capsule, with the dpa rate dependent on the axial location in MOTA.

With the exception of the below-core canister, the temperatures in MOTA capsules are actively controlled to ±5°C of the nominal target, although the nominal target temperatures varied a little from capsule to capsule. In the below-core capsule the temperatures are largely controlled by the inlet coolant temperature and the gamma heating level, and therefore can be calculated with rather small error.

The first irradiation sequence occurred in Cycle 11 of MOTA-2A for 2.58×10^{7} sec, and a subset of specimen packets was then removed. Other identical specimen packets continued in Cycle 12 of MOTA-2B for 1.71×10^{7} sec. The dose rates in the various capsules ranged from 8.9×10^{-9} to 1.7×10^{-6} dpa/sec. The dose levels attained by the specimens varied from 0.23 to 43.8 dpa in Cycle 11 and an additional 0.38 to 24.0 dpa in Cycle 12. Table 1 summarizes the irradiation.

Table 1 Irradiation conditions experienced by carbon- doped specimens in FFTF cycles 11 and 12 (MOTA-2A and MOTA-2B). Note that in three of the seven cases the specimens irradiated in both cycles did not experience completely identical conditions with single cycle packages.

Dose Rate, dpa/sec		Dose, dpa		Temperature, ˚C	
# 11	#12	#11	#11 & #12	#11	#11 & #12
1.7×10^{-6}	1.4×10^{-6}	43.8	67.8	427	408
7.8×10^{-7} *1	9.5×10^{-7}	20.0 *1	32.4	390	387
5.4×10^{-7}	8.4×10^{-7}	14.0	28.8	430	424
8.2×10^{-7}	-----	21.1	-----	430	-----
3.2×10^{-7} *2	3.5×10^{-7}	8.22 *2	13.1	373	373
3.1×10^{-7} *3	3.0×10^{-7}	8.05 *3	11.1	411	410
1.5×10^{-7}	1.3×10^{-7}	3.87	6.12	430	431
9.1×10^{-8}	2.1×10^{-7}	2.36	6.36	430	431
4.6×10^{-8}	4.2×10^{-8}	1.18	1.91	434	437
2.7×10^{-8}	6.6×10^{-8}	0.71	1.87	434	437
1.4×10^{-8}	1.4×10^{-8}	0.37	0.61	436	444
8.9×10^{-9}	2.2×10^{-8}	0.23	0.61	436	444

Note: The swelling data of the underlined irradiation conditions come from TEM observation, while the other are density measurements.

*1: 6.0×10^{-7} dpa/sec and 15.6 dpa for #11 in 2 cycle irradiation specimens
*2: 2.7×10^{-7} dpa/sec and 6.90 dpa for #11 in 2 cycle irradiation specimens
*3: 2.2×10^{-7} dpa/sec and 5.69 dpa for #11 in 2 cycle irradiation specimens

conditions for the twelve combinations of temperature, dpa and dpa rate

The starting and post-irradiation densities were measured using an immersion density technique known to be accurate to ±0.2 % change in density. In some cases it was not possible to clearly identify and retrieve all four specimens, but in general there were at least two identical specimens measured in each capsule. Determination of microstructural evolution in the specimens using a transmission electron microscope has been completed for the ternary and quaternary alloys, but has not yet been initiated for the carbon-doped alloy from MOTA-2B. An earlier study by Sekimura and Ishino addressed the microstructure of some of the MOTA-2A specimens, however (5). Both the current density change and earlier microscopy data are presented in Figure 2.

RESULTS

As shown in Figure 2, the swelling of the carbon-doped alloy at ~400ºC appears to show no obvious influence of the dpa rate. As was observed in the two simple undoped alloys, the range of swelling between identical specimens is relatively small, indicating the reproducibility of the swelling phenomenon. Surprisingly, the swelling of the twelve data ensemble of the carbon-doped alloy appears to be following a general, lower-swelling trend somewhat characteristic of the undoped alloys at the highest dpa rate.

Figure 3 shows that addition of 500 appm boron to the carbon-doped alloy reduced the swelling level somewhat, but the alloy in general retained its insensitivity to dpa rate.

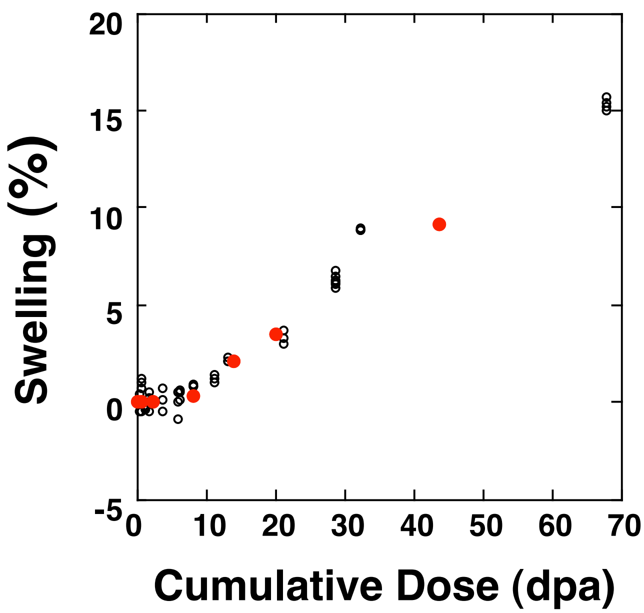

Figure 2. Swelling of simple model f.c.c. Fe-15Cr-16Ni-0.25Ti-0.05C in FFTF-MOTA at ~ 400˚C, showing that the swelling is relatively independent of dpa rate. The red data were obtained from irradiation in MOTA-2A only and the black data from irradiation in both MOTA-2A and 2B.

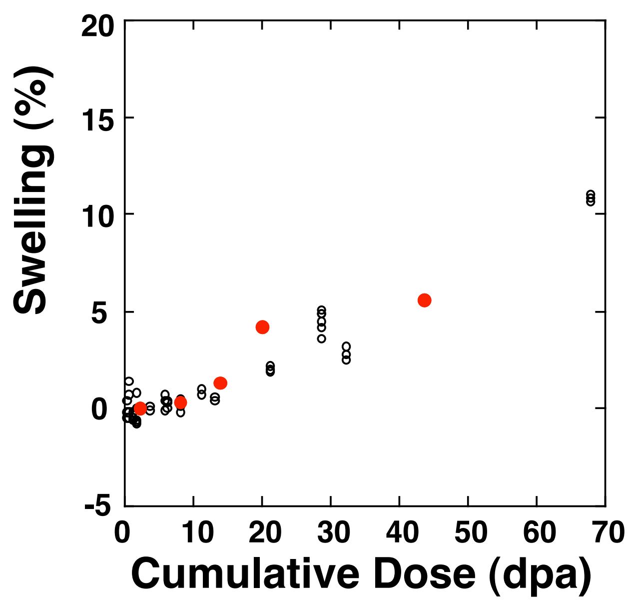

Figure 3. Swelling of simple model f.c.c. Fe-15Cr-16Ni-0.25Ti-0.05C-500 appm B in FFTF-MOTA at ~400ºC, showing that the swelling is reduced somewhat by boron addition but remains relatively independent of dpa rate. The red data were obtained from irradiation in MOTA-2A only and the black data from irradiation in both MOTA-2A and 2B

Figure 4 shows that most of the decrease in swelling observed at 500 appm boron was already attained at the 100 appm level. Beyond 500 appm there was a tendency to increase swelling, with the amount of the increase dependent on the dpa rate.

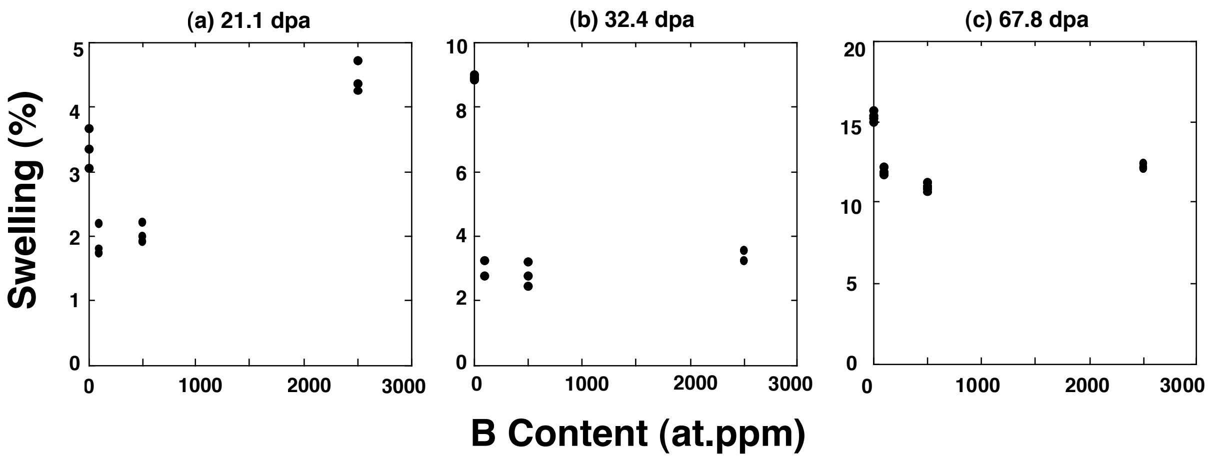

Figure 4. Swelling of simple model f.c.c. Fe-15Cr-16Ni-0.25Ti-0.05C with various levels of boron, as observed after irradiation in FFTF-MOTA at ~400ºC to three different dose rates and dose levels.

DISCUSSIONS

There are four major features of these results. First, carbon addition clearly suppresses the onset of swelling at ~400ºC, an effect not observed to result from the addition of Ti alone. Second, carbon addition suppresses the early termination of the transient regime, especially at lower dpa rates where the undoped alloys exhibited very short transient durations. Third, the carbon-induced suppression looks to be much less effective at higher dpa rates. Finally the addition of boron at 500 appm decreases the swelling somewhat but does not appear to change the insensitivity to dpa rate that occurs when carbon was added.

Currently, there are no microstructural observations for the MOTA-2B carbon-doped specimens to determine the origins of this surprising result. In the undoped alloys microstructural observations of both 2A and 2B specimens showed that the flux sensitivity of the transient regime arose primarily from the flux sensitivity of Frank loop evolution (1). Higher dpa rates produced higher densities of smaller loops which were more resistant to unfaulting and network formation, thereby delaying the development of a dislocation network. Attainment of a stable network was found to be coincident with the termination of the transient regime in these carbon-free alloys (1).

As shown in Figure 5, Sekimura and Ishino (5) earlier examined by microscopy another set of the MOTA-2A specimens at three irradiation temperatures including the ~400ºC specimens discussed in this report. Note that the influence of Ti addition is very small at ~400º but increases at higher temperatures, while carbon additions to the Ti-modified alloys suppress swelling at all temperatures in the range 400-600ºC.

Sekimura and Ishino did not address the effect of carbon on Frank loop characteristics at ~400ºC for MOTA-2A specimens, so we can not at this point invoke a specific role for carbon's possible influence to delay swelling via its action on the evolution on Frank loops. Microstructural examination of the MOTA-2B specimens in planned Phase 3 of this effort may allow the identification of a role for carbon.

Boron is well-known to interact with the precipitation of carbides and therefore may exert its influence on void swelling both directly or indirectly through its influence on carbon (6). With respect to boron's contribution it must be recognized that boron can play several roles, however, first as a chemical species and second as a source of transmutants helium and lithium produced by the $^{10}B(n, \_)\,^{7}Li$ reaction with thermal and epithermal neutrons.

Note, however, that ^{10}B comprises only 20% of natural boron. As shown in reference 1 some helium in these specimens is produced by high energy (n, _) reactions, primarily with nickel in these alloys, such that the boron-generated helium is additive to that generated in the base alloy.

In the core center positions from which the data shown in Figure 4 were attained, the impact of 100 appm boron is only a 30-40% increase in the helium generation rate which is already rather large. For example, at 67.8 dpa the boron contribution increased the helium from 15.5 appm generated by the base alloy to only 20 appm arising from the 100 appm boron addition.

Since the primary impact of boron was attained by 100 appm boron and the increase in helium is not large, it is concluded by the authors that the primary effect of boron is probably chemical in nature and arises from its influence on carbon and its distribution in the alloy.

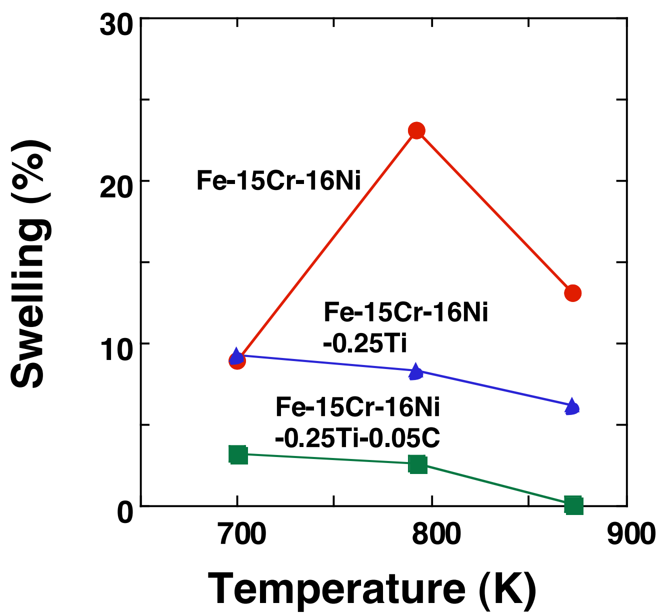

Figure 5 Cavity volume fraction determined by microscopy of three simple austenitic alloys after irradiation in FFTF-MOTA-2A at three different temperatures (5). Note that addition of titanium has no effect at 400˚C, but carbon addition has a strong decreasing effect on swelling.

CONCLUSIONS

The addition of 0.05% carbon to Fe-15Cr-16Ni-0.25Ti leads to a reduction of neutron-induced swelling at ~400ºC, with the reduction being largest when the irradiation is conducted at relatively low dpa rates. Another consequence of carbon addition is that the strong influence of the dpa rate observed in the carbon-free alloy completely disappears in the carbon-doped alloy. The microstructural reasons for this behavior are not yet clear.
Boron additions modify the swelling somewhat but appear to arise from the influence of boron as a chemical species rather than as a source of helium.

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REFERENCES

1. T. OKITA, N. SEKIMURA, F. A. GARNER, L. R. GREENWOOD, W. G. WOLFER and Y. ISOBE, "Neutron-Induced Microstructural Evolution of Fe-15Cr-16Ni Alloys at ~ 400°C During Neutron Irradiation in the FFTF Fast Reactor", *10th International Conference on Environmental Degradation of Materials in Nuclear Power Systems – Water Reactors, issued on CD format, no page numbers, 2001.* Also in Fusion Materials Semiannual Progress Report for Period Ending June 30,**vol. 30***, pp. 148, 2001*
2. J. L. SERAN and J. M. DUPOUY, "The Swelling of Solution Annealed 316 Cladding in Rapsodie and Phenix", *Effects of Radiation on Materials: 11th International Symposium ASTM STP* **782***, pp. 5, 1982*
3. J. L. SERAN and J. M. DUPOUY, "Effect of Time and Dose Rate on the Swelling of 316 Cladding in Phenix", *Proceedings Conference on Dimensional Stability and Mechanical Behavior of Irradiated Metals and Alloys, Vol. 1, BNES, pp. 25, 1983*
4. G. M. BOND, B. H. SENCER, F. A. GARNER, M. L. HAMILTON, T. R. ALLEN and D. L. PORTER, "Void Swelling of Annealed 304 Stainless Steel at ~370-385°C and PWR-Relevant Displacement Rates", *9th International Conference on Environmental Degradation of Materials in Nuclear Power Systems – Water Reactors, pp. 1045, 1999*
5. N. SEKIMURA and S. ISHINO, "The Effect of Titanium Addition on Microstructural Evolution in Austenitic Steel Irradiated with Fast Neutrons in FFTF", *Journal. of Nuclear Materials* **179-181** *pp. 542 1991.*
6. F. A. GARNER, "*Irradiation Performance of Cladding and Structural Steels in Liquid Metal Reactors,*" Vol. 10A Chapter 6. pp. 419-543. Materials Science and Technology: A Comprehensive Treatment, VCH Publishers, 1994,